

Development of a selective separation method for element 107 – bohrium – based on fast liquid-liquid extractions

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Abstract

Liquid-liquid extraction has been used to develop a fast separation procedure of the homologues of element 107 (bohrium, Bh) from other reaction products, which can simultaneously be produced in the heavy ion reactions used for transactinide production. Alpha-hydroxyisobutyric acid has been used as complexing agent in the aqueous phase solution. The organic phase solution consisted of trioctylamine in Shellsol T. A quantitative extraction of technetium and rhenium has been observed in the pH range from 1.5 to 5.5, with a good separation from most other studied elements, except for iodine, which has been extracted up to 50 % and niobium, which only can be separated from the group VII elements at high pH-values.

Introduction

Knowledge about the chemical properties of transactinide elements allows to determine their position within the periodic table of the elements. If they behave similar to their lighter homologues they can simply be placed under the corresponding group. If their chemical behavior is influenced by relativistic effects, positioning within the table of elements may be more difficult.

Transactinide elements are studied using different chemical separation methods including column separation, gas chromatography and liquid-liquid extraction. Usually only a few atoms of these elements are produced during an accelerator experiment and the half-lives of these elements are in the range of seconds to minutes. Therefore the separation method used has to be very fast, highly selective for the element of interest, very efficient and also the detection method should be capable to measure single atoms with high accuracy and efficiency.

The SISAK (Short-lived Isotopes studied by the AKUFVE technique) [1] uses fast rotating centrifuges to perform continuous liquid-liquid extractions. In the past, this system has been coupled with an on-line alpha-liquid scintillation system to be able to continuously measure alpha-decaying transactinides.

Presently new developments are taking place to further improve the performance of the SISAK system: to overcome problems with high overall activity and resulting detection difficulties a pre-separation of the produced transactinide with a gas-filled separator is investigated. First experiments have shown promising results [2]. Secondly, the design of a so-called 'micro-SISAK' system is going on, which should reduce the total amount of aqueous and organic solutions used for the separation [3] and also will allow operation of the system at lower flow-rates. In this case, detection of the extracted nuclides using semi-conductor detectors is also investigated.

However, liquid-liquid extraction still remains the main principle on which the separation is based. Such a liquid-liquid extraction procedure is usually investigated by performing experiments with the lighter homologues of the transactinide of interest as well as some neighbouring elements. In the case of bohrium (element 107), technetium and rhenium are used as homologues and are separated from group 4, 5 and 6 elements, as well as some di- and tri-valent metals and one actinide element.

Experimental

Radionuclides

Table 1 shows the radionuclides used for developing the extraction procedure. If they were not commercially available, production procedures were developed to obtain non-carrier-added radionuclides. These production procedures were usually based on neutron irradiation of certain compounds at the HOR research reactor of the Interfaculty Reactor Institute in Delft, The Netherlands. Ideally, (n,p)- and (n,α)- reactions were used to produce the short-lived nuclides. Examples are the production of non-carrier-added ^{64}Cu and ^{58}Co . Other nuclides were produced by beta-decay of the corresponding mother nuclide: ^{95}Nb and ^{105}Rh were produced in this way. Before the radionuclides were used for the extraction experiments, they were chemically separated from their original matrix or mother nuclides.

Table 1. Radionuclides used to develop an extraction procedure for bohrium and their half-lives.

Radionuclide	$t_{1/2}$
$^{99\text{m}}\text{Tc}$	6.02 h
^{186}Re	3.78 d
^{188}Re	16.98 h
^{95}Nb	31.15 d
^{105}Rh	1.47 d
^{103}Ru	39.35 d
^{239}Np	2.36 d
^{131}I	8.04 d
^{65}Zn	244 d
^{64}Cu	12.71 h
^{58}Co	70.78 d

Extractions

The aqueous solution consisted of 1 M α-hydroxyisobutyric acid, 0.01 M potassium bromate to ensure the +VII oxidation state of the elements, and 0.22 M dodecanol to improve the phase separation. The pH-value was varied using either hydrochloric acid or ammonia. The organic solution consisted of 2 % vol trioctylamine (TOA) in Shellsol T. Liquid-liquid extractions were performed in polyethylene tubes: the tubes were sealed at one end and 0.5 ml of the two phases including the nuclide of interest were added in the tube. After sealing the other end of the tube it was shaken 1 minute using a Vortex apparatus. The phases were separated by centrifugation and an aliquot was removed from both phases

and measured using Ge(Li) semiconductor detector. Extractions were carried out three times and the average value is given for the extraction yield.

Results and Discussion

Figure 1 shows the results obtained for extraction of the above mentioned nuclides: Tc and Re are extracted quantitatively into the organic phase throughout the whole pH range from 1.6 to 5.0. Niobium as a homologue of element 105 is extracted in high yields at lower pH values, but the yield decreases for higher pH values until it is extracted less than 5 % at pH values around 5. This result is in agreement with earlier experiments [4]. Iodine is extracted with a yield of about 50 % throughout the whole pH range. Cobalt, copper, neptunium and zinc are not extracted at all. The extraction yields of rhodium and ruthenium increase slightly at higher pH values and reach a maximum yield of about 17 % and 30 % and pH values 5.5 and 4.5, respectively.

First results obtained with molybdenum show negligible extraction yields and are not yet included into figure 1.

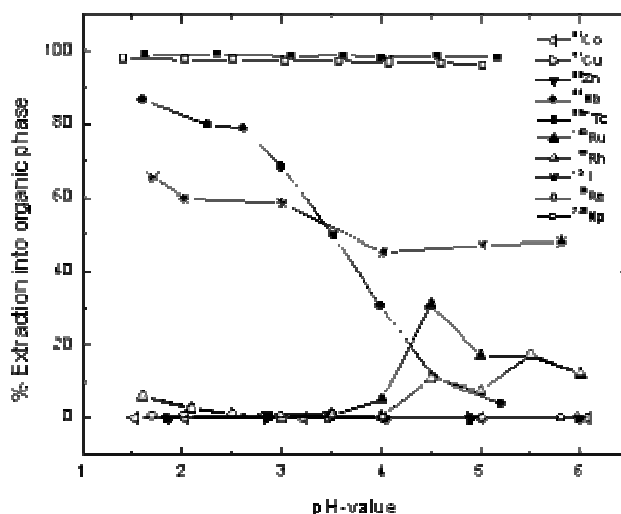


Figure 1. Extraction yields for different nuclides. Organic phase: 2 % Vol trioctylamine in Shellsol, aqueous phase: 1 M alpha-hydroxyisobutyric acid, 0.01 M potassium bromate, 0.22 M dodecanol.

The results suggest that this extraction system using alpha-hydroxyisobutyric acid may be used at pH values above 5 for a quantitative separation of element 107 and its homologues from other reaction products.

Future work

Not only molybdenum but also tungsten isotopes will be used in future experiments to ensure the selective separation of group 6 from group 7 elements. Addition of a small amount of H_2O_2 to the aqueous phase should result in a decreased extraction yield of niobium and will also be investigated. Furthermore, on-line experiments with even shorter lived isotopes are necessary to ensure that the extraction kinetics is indeed fast enough for future continuous separations using the SISAK system.

Also, if on-line alpha-liquid-scintillation counting will be used as detection method for the transactinide element, the organic phase should consist of an aromatic solvent, such as toluene. Although earlier experiments suggest that the above mentioned extractions are in general possible with aromatic solvents [5], some experiments have to be carried out to ensure identical extraction yields as when using Shellsol.

References

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